Optical Pumping and Interaction of Atoms with the Electromagnetic Field

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Optical pumping can be briefly described as the transfer of angular momentum from polarized photons to atoms. It provides very sensitive optical detection of any change in the angular state of the atom, resulting from RF transitions between Zeeman sublevels, relaxation processes, etc. ...

The principal applications of optical pumping may be divided into 3 parts:

—spectroscopic measurements
—study of relaxation processes and collisions
—study of the interaction of oriented atoms with the e.m. field.

This course will describe some effects related to the third part.†

We will study how oriented atoms are perturbed when they are irradiated by light or RF photons. When the frequency of the impinging photon is equal to the Bohr frequency of an atomic transition, the atom absorbs it. Energy is conserved during such a transition and the process is called a *real* absorption. If energy cannot be conserved during the transition, the interaction is described in terms of *virtual* absorption and reemission of photons by the atom. The purpose of these lectures is to describe some effects of these real or virtual absorption of optical and RF photons.

The first two lectures will be devoted to the interaction with optical photons. It will be shown, first theoretically then experimentally, how atomic sublevels in the ground state are broadened and shifted by irradiation with resonant or near resonant optical photons. The last two lectures will describe the interactions with RF photons and will essentially be devoted to the study

* Lecture Notes taken by J. Dupont-Roc, D. Ostrowsky and N. Polonsky.

† For a detailed description of the basic principles, experimental techniques and the other applications of optical pumping, see references[1].

of higher order processes involving several quanta. The possibility of cancelling the Landé factor of an atom by interaction with a RF field will also be described. The understanding of all these effects requires the knowledge of crossing and anticrossing of energy levels. So this concept will be first described in the third lecture.

I. Interaction of Atoms with Optical Photons

Introduction

When atoms, in the ground state, are irradiated with resonant or quasi resonant optical photons, the problems to be solved are the following:

How are the atoms excited?
What do they do in the excited state?
How do they fall back to the ground state?

These problems have been studied in great detail in references.[2] In this course we will focus our attention only on the first step (excitation process) and use a different approach based on the resolvant formalism.[3,4] As this formalism will prove to be useful also for the other chapters of this course it will be first briefly reviewed (§ A). We will then apply it to the problem of the light shifts (§ B). We will finally describe the experimental results some of which have been obtained very recently (§ C).

A. Resolvant formalism

(1) *Definition.* The resolvant $G(z)$ of the Hamiltonian $\mathscr{H}$ is by definition:

$$G(z) = \frac{1}{z - \mathscr{H}} \tag{I.1}$$

$G(z)$ is simply related to the evolution operator $U(t) = e^{-i\mathscr{H}t}$:

$$U(t) = \frac{1}{2\pi i} \int_C e^{-izt}\, G(z)\, dz \tag{I.2}$$

where C is the following contour of the complex plane

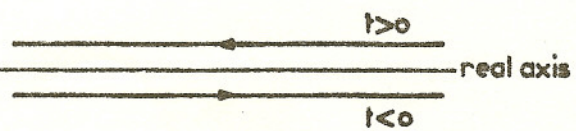


Fig. 1

It may be easily shown that $G(z)$ is an analytic function on the complex z plane except eventually on the real axis where poles and cuts corresponding to the discrete and continuous eigenvalues of $\mathscr{H}$ may be found.

For $t > 0$ ($t < 0$), only the part of the contour C above (below) the real axis gives a non zero contribution.

—The matrix elements of $G(z)$ are often more easily evaluated than those of $U(t)$. This explains the importance of $G(z)$. If $\mathscr{H} = \mathscr{H}_0 + V$, and if $|a\rangle$ is an eigenstate of $\mathscr{H}_0$ evaluation of $\langle a| G(z) |a\rangle$ gives, by (I.2), the probability amplitude that, the system being in state $|a\rangle$ at $t = 0$, it remains in the same state at time t. This is important for decay problems. Evaluation of $\langle b| G(z) |a\rangle$ leads also to the transition amplitude from $|a\rangle$ to another state $|b\rangle$ of $\mathscr{H}_0$ under the effect of V.

—It is usually not necessary to calculate all the matrix elements of G. Depending on the problem to be solved, only a few of them must be evaluated. So we will now derive an explicit expression for the projection, $\bar{G}(z)$, of $G(z)$ into a subspace, ε†, of the Hilbert space. If P (and Q) are the projections onto (out of) ε

$$P^2 = P \qquad P + Q = 1 \qquad PQ = 0$$

$$Q^2 = Q \qquad [P, \mathscr{H}_0] = 0 \qquad [Q, \mathscr{H}_0] = 0$$

$$\bar{G}(z) = PG(z)P \tag{I.3}$$

and in general way for any operator A:

$$\bar{A} = PAP$$

$\bar{G}$ and $\bar{A}$ operate only inside ε.

(2) *Explicit expression for* $\bar{G}(z)$. Using the identity:

$$\frac{1}{A} - \frac{1}{B} = \frac{1}{A}(B - A)\frac{1}{B}$$

we obtain:

$$G = G_0 + G_0 V G \tag{I.4}$$

where G_0 is the resolvant of the Hamiltonian $\mathscr{H}_0$:

$$G_0 = \frac{1}{z - \mathscr{H}_0}$$

From (I.4) it is easy to compute $\bar{G} = PGP$

$$\bar{G} = \frac{P}{z - \mathscr{H}_0} + \frac{P}{z - \mathscr{H}_0} VGP$$

† spanned by eigenstates of $\mathscr{H}_0$.

Using the equality $P + Q = 1$, we have also:

$$G = \frac{P}{z - \mathscr{H}_0} + \frac{1}{z - \mathscr{H}_0} VG + \frac{P}{z - \mathscr{H}_0} VQGP. \tag{I.5}$$

On the other hand, from the definition of G:

$$(z - \mathscr{H}_0 - V) G = 1$$

we get

$$Q (z - \mathscr{H}_0 - V) (P + Q) GP = 0$$

or

$$[z - \mathscr{H}_0 - QVQ] QGP = QVPG \tag{I.6}$$

Replacing in (I.5) QGP by this new expression, we get:

$$G = \frac{P}{z - \mathscr{H}_0} + \frac{1}{z - \mathscr{H}_0} VG + \frac{P}{z - \mathscr{H}_0} V \frac{Q}{z - \mathscr{H}_0 - QVQ} VPG$$

or

$$\left[z - \mathscr{H}_0 - V - PV \frac{Q}{z - \mathscr{H}_0 - QVQ} VP \right] G = P$$

We finally obtain:

$$G(z) = \frac{1}{z - \bar{\mathscr{H}}_0 - \bar{R}(z)} \tag{I.7}$$

where

$$\bar{R}(z) = PVP + PV \frac{Q}{z - \mathscr{H}_0 - QVQ} VP \tag{I.8}$$

$\bar{R}(z)$ can be expanded for small V:

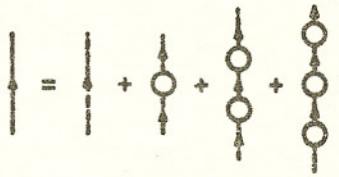
$$\bar{R}(z) = PVP + PV \frac{Q}{z - \mathscr{H}_0} VP + PV \frac{Q}{z - \mathscr{H}_0} V \frac{Q}{z - \mathscr{H}_0} VP + \cdots \tag{I.9}$$

The structure of the matrix elements of $\bar{R}(z)$ is simply the product of matrix elements of V and energy denominators. But, due to the presence of Q, all the intermediate states must be outside ε. In other words, the diagrams representing $\bar{R}(z)$ must be irreductible, in the sense that internal and external lines do not correspond to the same subspace.

—Equation (I.7) is rigorous. If $\bar{R}(z)$ is small compared to $\bar{\mathscr{H}}_0$ one can develop (I.7) in a power series of $\bar{R}(z)$:

$$G = G_0 + G_0 \bar{R} G_0 + G_0 \bar{R} G_0 \bar{R} G_0 + \cdots \tag{I.10}$$

and represent the development so obtained by diagrams:



(I.11)

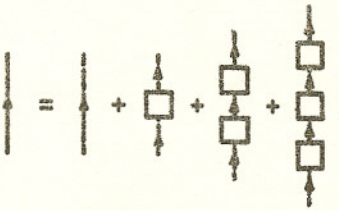
the full line (true propagator) being associated to $\bar{G}(z)$, the dotted line (free propagator) to G_0, and the circles to $\bar{R}(z)$.

Up to now, no approximation has been made. If V is small, we can replace in (I.7) $\bar{R}(z)$ by an approximate value, $\bar{R}'(z)$, obtained for example by keeping only the first two terms of equation (I.9):

$$\bar{G}(z) = \frac{\cdot\ 1}{z - \mathscr{H}_0 - \bar{R}'(z)} \qquad \text{(I.10 bis)}$$

$$= G_0 + G_0\bar{R}'G_0 + G_0\bar{R}'G_0\bar{R}'G_0 + \cdots$$

This amounts to replacing in (I.11) all the circles by the diagrams associated with the approximate expression $\bar{R}'(z)$ and represented by squares in (I.12).



(I.12)

But it must be emphasized that this is much better than an ordinary perturbation treatment where we would have kept only a *finite* number of the diagrams of (I.11). By making an approximation only on $\bar{R}(z)$ in the expression (I.7), and so by keeping all the terms of the expression (I.10 bis), we do in fact sum an entire class of diagrams which are the most important and we keep terms with arbitrarily high powers of V as it appears in (I.12). This is of great interest in many problems such as the decay of excited atomic states which we discuss in the next paragraph as an application of the resolvant formalism.

(3) *Decay of an atomic excited state.* Consider a two-level atom (Fig. 2), coupled by the hamiltonian V to the electromagnetic field. We study the decay of the excited state. The hamiltonian for the problem is $\mathscr{H} = \mathscr{H}_0 + V$ where $\mathscr{H}_0$ represents the energy of the atom and of the free radiation field.

To the first order, V couples only the two states $|e, o\rangle$ (excited state with no photon) and $|g\vec{k}\vec{\varepsilon}_\lambda\rangle$ (ground state with a photon of momentum $\vec{k}$ and polarization $\vec{\varepsilon}_\lambda$).

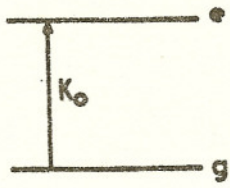


Fig. 2

The two stationary states for the problem are $|g, \vec{k}, \vec{\varepsilon}_\lambda\rangle$ and $|e, o\rangle \equiv |a\rangle$.
We have $\langle a| V |a\rangle = 0$ and $\langle a| V |g\vec{k}\vec{\varepsilon}_\lambda\rangle \neq 0$.
Taking the zero of energy as the ground state, we have:

$$\mathscr{H}_0 |a\rangle = k_0 |a\rangle \tag{I.13}$$

To study the decay of $|a\rangle$, we shall consider a one dimensional subspace ε, subtended by $|a\rangle$ and calculate the matrix element $\overline{G}_a(z) = \langle a| \overline{G} |a\rangle$ using (I.7) and (I.13); we get:

$$\overline{G}_a(z) = \frac{1}{z - k_0 - \overline{R}_a(z)} \tag{I.14}$$

We shall approximate $\overline{R}_a(z)$ by the first two terms of its expansion in powers of V:

$$\overline{R}'_a(z) = \langle a |V| a\rangle + \left\langle a \left| PV \frac{Q}{z - H_0} VP \right| a \right\rangle$$

or

$$\overline{R}'_a(z) = \sum_k \sum_\lambda \frac{|\langle a |V| g\vec{k}\vec{\varepsilon}_\lambda\rangle|^2}{z - k} \tag{I.15}$$

If $\overline{R}'_a(z)$ is small and smooth, the only values of z for which $\overline{G}_a(z)$ is important are near $z = k_0$. We approximate† $\overline{G}_a(z)$ in the upper half plane near the real axis by

$$\overline{G}_a(z) \simeq \frac{1}{z - k_0 - \overline{R}'_a(k_0 + i\varepsilon)}$$

So $\overline{R}'_a$ may be written in the following way:

$$\overline{R}'_a = \sum_k \sum_\lambda \frac{\langle |a| V |g\vec{k}\vec{\varepsilon}_\lambda\rangle|^2}{k_0 - k + i\varepsilon}$$

† A more correct and rigorous treatment based on analytic continuation of $\overline{G}_a(z)$ is presented in references[4].

or, replacing the summation over $\vec{k}$ by integrations over the length and the solid angle Ω of $\vec{k}$:

$$\overline{R'_a} = \sum_{\lambda} \int k^2 \, d\Omega \, dk \, \frac{|\langle a|V|g\vec{k}\vec{e}_\lambda\rangle|^2}{k_0 - k + i\varepsilon} = \Delta E - i\frac{\Gamma}{2}$$

where

$$\begin{cases} \Delta E = \int \mathcal{P}\left(\dfrac{1}{k_0 - k}\right) k^2 \, dk \sum_{\lambda} \int d\Omega \, |\langle a|V|g\vec{k}\vec{e}_\lambda\rangle|^2 \\ \Gamma = 2\pi \int \delta(k_0 - k)\, k^2 \, dk \sum_{\lambda} \int d\Omega \, |\langle a|V|g\vec{k}\vec{e}_\lambda\rangle|^2 \end{cases}$$

The final expression of $\overline{G_a}(z)$ is thus:

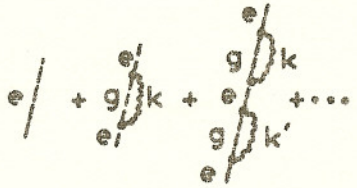
$$\overline{G_a}(z) = \frac{1}{z - k_0 - \Delta E + i\dfrac{\Gamma}{2}} \tag{I.16}$$

From (I.16), one gets easily

$$\langle a|U(t)|a\rangle = e^{-ik_0 t} e^{-i\Delta E t} e^{-(\Gamma/2)t} \tag{I.17}$$

We have then found the shift ΔE and the natural width Γ of the excited state.

In this treatment we have made a partial summation of an infinite set of diagrams:



neglecting diagrams of the type:

This means we have approximated $e^{-\frac{(\Gamma)_{true}}{2}t}$ by

$$1 - \frac{\Gamma}{2}t + \frac{\left(-\dfrac{\Gamma}{2}\right)^2 t^2}{2!} + \frac{\left(-\dfrac{\Gamma}{2}\right)^3 t^3}{3!} + \cdots$$

We have done an infinite summation to find the exponential behavior, the only approximation being associated with the value of Γ which differs slightly from $(\Gamma)_{true}$.

B. Light shifts

We shall now use the resolvant formalism to study the evolution of an atom irradiated by a light beam.

In a first step, we shall consider a simple atomic model with no Zeeman structure, and give a physical interpretation of the results obtained.

In the second part we shall generalize the theoretical formulas to the more complicated case of an atom with a Zeeman structure. We shall then give some experimental evidence for the predicted effects.

(1) *Simple model. No Zeeman structure.* We consider an atom which has a ground state and an excited state separated by the energy k_0. It is irradiated by N optical photons $\vec{k}_1, \ldots \vec{k}_i, \ldots \vec{k}_N$ whose energy distribution is given by the line shape $u(k)$ (Fig. 3). We shall call $\vec{k}$ the center and Δ the width of $u(k)$.

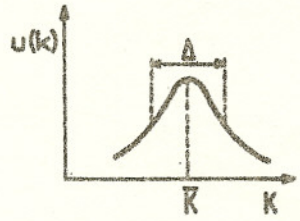


Fig. 3

All the N photons have the same polarization $\vec{\varepsilon}_{\lambda_0}$.

We consider the two following states of the total system atom + radiation field:

$|g\rangle$: atom in the ground state in the presence of the N photons.

$|e, -\vec{k}_i\rangle$: atom in the excited state; the photon $\vec{k}_i$ having been absorbed.

$|g\rangle$ and $|e, -\vec{k}_i\rangle$ are eigenstates of the unperturbed hamiltonian $\mathscr{H}_0$ with eigenvalues E_0 and $E_0 + k_0 - k_i$ and are coupled by the interaction hamiltonian V. V couples also $|g\rangle$ to other states with $N+1$ photons: $|e, +\vec{k}\rangle$ (photon k emitted) but we shall neglect them because of the large energy difference between $|g\rangle$ and $|e, +\vec{k}\rangle$.

The system at time $t = 0$ is in the state $|g\rangle$; we look for the probability that it remains in the same state at time t. We thus have to compute

$$G_g(z) = \langle g\,|G(z)|\,g\rangle$$

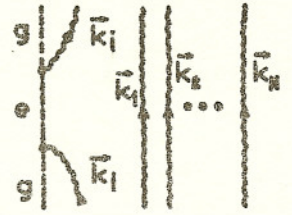
As in equation (I.14), we write:

$$\overline{G}_g(z) = \frac{1}{z - E_0 - \overline{R}_g(z)} \tag{I.18}$$

We approximate now $\overline{R_g}(z)$ by the first non-zero term in its expansion in powers of V:

$$\overline{R'_g}(z) = \sum_i \frac{|\langle g|V|e, -k_i\rangle|^2}{z - E_0 - k_0 + k_i} \tag{I.19}$$

The basic diagram associated to $\overline{R'_g}$ is the following

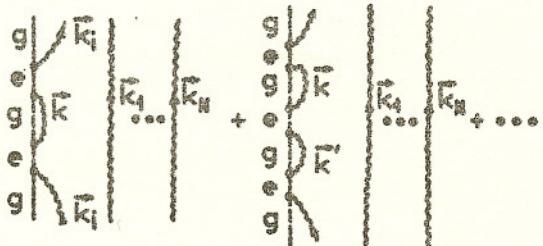


In order to improve slightly the expression (I.19), we take into account spontaneous emission during the intermediate excited state e, by replacing the free propagator $\dfrac{1}{z - E_0 - k_0 + k_i}$ of the excited state by the true propagator of this state. If the induced emission due to $\vec{k}_1, \vec{k}_2, \ldots, \vec{k}_N$ is negligible in comparison to the spontaneous emission, which is the case for ordinary light sources, we can calculate this propagator as if the atom was isolated, i.e., without incident light beam: according to § I-A-3 we have to replace k_0 by $k_0 + \Delta E - i\dfrac{\Gamma}{2}$ where ΔE and Γ are the shift and the natural width of the excited state. So we get for the improved approximation of $\overline{R'_g}$

$$\overline{\overline{R'_g}}(z) = \sum_i \frac{|\langle g|V|e, -\vec{k}_i\rangle|^2}{z - E_0 - \vec{k}_0 + i\dfrac{\Gamma}{2} + k_i} \tag{I.20}$$

with $\vec{k}_0 = k_0 + \Delta E$.

Diagrammatically one gets (I.20) by adding to the basic diagram associated with $\overline{R'_g}$, an infinite number of other diagrams which were omitted in the approximation (I.19)



(we neglect also the possibility of coincidences between $\vec{k}$, $\vec{k}'$ and $\vec{k}_1 \cdots \vec{k}_N$ when we simply replace k_0 by $k_0 - i\frac{\Gamma}{2}$).

Now, as in section § I-A-3, we notice that in the expression (I.18) of $\bar{G}_g(z)$ one can neglect $\bar{R}_g(z)$ except when z is nearly equal to E_0. We shall therefore approximate $\bar{R}''(z)$ by $\bar{R}''(E_0 + i\varepsilon)$:

$$\overline{R''_g}(E_0 + i\varepsilon) = \sum_l \frac{|\langle g|V|e, -\vec{k}_l\rangle|^2}{k_l - k_0 + i\varepsilon + i\frac{\Gamma}{2}} \tag{I.20'}$$

We separate now the angular part of the matrix element of V

$$|\langle g|V|e, -\vec{k}_l\rangle|^2 = |A_{k_l}|^2 \, |\langle g|\vec{e}_{\lambda_0}\vec{D}|e\rangle|^2$$

$\vec{D}$ is the angular part of the electric dipole operator.

A_{k_l} contains the radial part.

We replace the summation over all the photons $\vec{k}_l$ by an integration over k, weighted by the line shape $u(k)$ and we get

$$\overline{R''_g}(E_0 + i\varepsilon) = p\left(\Delta E' - i\frac{\Gamma'}{2}\right) \tag{I.21}$$

where

$$\begin{cases} p = |\langle g|\vec{e}_{\lambda_0}\vec{D}|e\rangle|^2 & \text{(I.22)} \\ \Delta E' = \int |A_k|^2 \, u(k) \dfrac{k - k_0}{(k - k_0)^2 + \dfrac{\Gamma^2}{4}} \, dk & \text{(I.23)} \\ \dfrac{\Gamma'}{2} = \int u(k) \, |A_k|^2 \dfrac{\dfrac{\Gamma}{2}}{(k - k_0)^2 + \dfrac{\Gamma^2}{4}} \, dk & \text{(I.24)} \end{cases}$$

Finally we have

$$\bar{G}_g(z) = \frac{1}{z - E_0 - p\left(\Delta E' - i\frac{\Gamma'}{2}\right)} \tag{I.25}$$

from which we deduce

$$U_g(t) = e^{-i(E_0 + p\Delta E')t} \, e^{-p(\Gamma'/2)t}$$

The interpretation of $\Delta E'$ and Γ' follows immediately. Under the effect of the light irradiation, the ground state has now a finite life time $1/p\Gamma'$, and an energy which is shifted from E_0 to $E_0 + p\,\Delta E'$ (light shift).

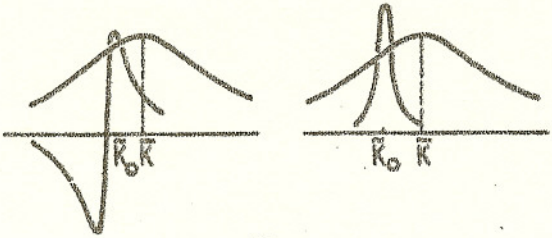


Fig. 4

We see from (I.23) and (I.24) that to get $\Delta E'$ or Γ', we must multiply $w(k)$ by the atomic dispersion or absorption curve, and integrate over k from 0 to ∞ (Fig. 4)

Let us now study how Γ' and $\Delta E'$ vary with $\bar{k} - \bar{k}_0$. From (I.23) and (I.24) one can plot the theoretical curves (Fig. 5). The value of $\bar{k} - \bar{k}_0$ corresponding to the maximum of $\Delta E'$ is of the order of Δ.

Furthermore, Fig. 4 shows that Γ' depends only on resonant photons whose energy lies between $k_0 - \Gamma$ and $k_0 + \Gamma$; on the contrary, $\Delta E'$ depends only on non resonant photons which have an energy either larger than $k_0 + \Gamma$ or less than $k_0 - \Gamma$. This leads us to consider two types of transitions.

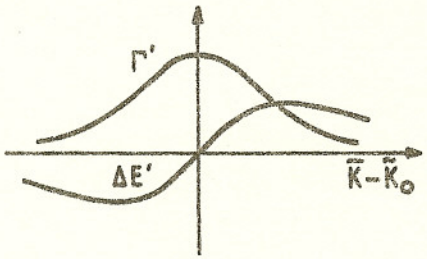


Fig. 5

If $\bar{k} - \bar{k}_0$ is small enough, effective absorptions of photons are possible, and those *real transitions* give a finite life time $1/p\Gamma'$ to the ground state.

On the other hand, if $\bar{k} - \bar{k}_0$ is large, the photons can only be absorbed during a very short time of the order of $\dfrac{1}{\bar{k} - \bar{k}_0}$: through those *virtual*

absorptions, the wave functions of the ground and excited states are mixed, and consequently the energy of those states are shifted.

What are the effects of these transitions on the photons?

In real transitions, photons disappear. This is absorption.

In virtual transitions, during a very short time, photons are absorbed and do not propagate. This is the basis of anomalous dispersion.

This can be summarized in a short table

Effects on the photons		*Effects on the atoms*
absorption	← real transition →	finite life time of the atomic ground state.
anomalous dispersion	← virtual transition →	shift of the ground state energy level

We are now ready to study the more general and usual case where both the excited and the ground states have a Zeeman structure.

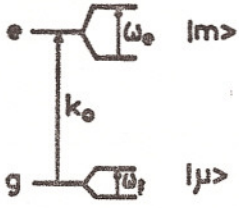


Fig. 6

(2) *More exact model. Zeeman structure.* Let us call $|\mu\rangle$ and $|m\rangle$ the atomic Zeeman sublevels of the ground and excited states in a steady magnetic field H_0 (Fig. 6)

If γ_e and γ_g are the gyromagnetic ratios of the excited and ground states, we call

$$\omega_e = \gamma_e H_0$$

$$\omega_g = \gamma_g H_0$$

We assume, as is generally the case in an optical pumping experiment, that $\omega_e, \omega_g \ll \Delta$, the width of the line shape, $u(k)$, of the incident beam.

The calculations are very similar to those of § I-B-1. For this particular problem, we consider a subspace ε subtended by all the substates $|\mu\rangle$. The projector P on ε is: $P = \sum_{\mu} |\mu\rangle \langle\mu|$ and we have to compute $G = PGP$.

As in § I-B-1, we obtain

$$G = \frac{1}{z - \bar{H}_0 - \bar{R}''(E_0 + i\varepsilon)} \tag{I.26}$$

where $\bar{R}''$ instead of being a number as in (I.20′) is now an operator which acts on the ground state multiplicity and which represents the effective hamiltonian $\mathscr{H}_L$ describing the effect of the light beam on the atomic ground state.

The generalization of (I.21) is

$$\bar{R}''(E_0 + i\varepsilon) = \mathscr{H}_L = \left(\Delta E' - i\frac{\Gamma'}{2}\right) A$$

where $\Delta E'$ and Γ' have been defined in (I.23) and (I.24) and where A is a matrix which generalizes (I.22)

$$\langle \mu | A | \mu' \rangle = A_{\mu\mu'} = \sum_m \langle \mu | \vec{\varepsilon}^*_{\lambda_0} \vec{D} | m \rangle \langle m | \vec{\varepsilon}_{\lambda_0} \vec{D} | \mu' \rangle$$

As A is hermitian, it has real eigenvalues. Let us call $|\alpha\rangle$ the eigenstate corresponding to the eigenvalue p_α:

$$A|\alpha\rangle = p_\alpha |\alpha\rangle.$$

According to (I.26) the state $|\alpha\rangle$ has a width $p_\alpha \Gamma'$ and an energy shift $p_\alpha \Delta E'$.

In general, the p'_αs are not all equal. So the various ground state sublevels are shifted differently. This is important for the experimental observation of the light shifts as we shall see later on.

Let us take, as an example, the simple case of ^{199}Hg which has only two Zeeman sublevels in the ground state. The two dimension matrix A may be expanded as:

$$A = a_0 I + \sum_{i=x,y,z} a_i \sigma_i$$

where I is the unit matrix and the σ_i's are the Pauli matrices.

This means that, if the incident beam contains mostly non resonant photons, $\mathscr{H}_L$ is approximately equal to $\Delta E' A$ and is similar to a Zeeman hamiltonian: the effect of the beam on the atoms is equivalent to that of a fictitious magnetic field $\vec{H}_f$†.

In a real experiment, the atom is also acted on by a real magnetic field H_0 parallel to Oz. Let us call $\mathscr{H}_z$ the real Zeeman hamiltonian. The total hamiltonian

† By symmetry considerations, one can easily show that $\vec{H}_f$ is parallel to the incident beam.

of the ground state is then:

$$\mathcal{H} = \mathcal{H}_L + \mathcal{H}_z$$

and we have the two relations:

$$\begin{cases} \mathcal{H}_L |\alpha\rangle = p_\alpha \left(\Delta E' - i \dfrac{\Gamma'}{2} \right) |\alpha\rangle \\ \mathcal{H}_z |\mu\rangle = \mu \omega_g |\mu\rangle \end{cases}$$

To study the energy diagram of the system, let us consider the two extreme cases:

a) $\mathcal{H}_L \gg \mathcal{H}_z$: the eigenstates of the system are the $|\alpha\rangle$ states which have different energies: the Zeeman degeneracy is removed. The splitting at $H_0 = 0$ is due, of course, to $\mathcal{H}_L$.

b) $\mathcal{H}_L \ll \mathcal{H}_z$: the effect of the light may be treated as a small perturbation. According to perturbation theory, the eigenstates $|\mu\rangle$ of the system have an energy which differs slightly from $\mu\omega_g$ by the amount

$$\begin{aligned} \Delta E_\mu &= \langle \mu | \mathcal{H}_L | \mu \rangle \\ &= \left(\Delta E' - i \frac{\Gamma'}{2} \right) A_{\mu\mu} \end{aligned}$$

The two energy levels of the system are plotted as functions of the magnetic field H_0 in Fig. 7 (in the particular case where the light beam is perpendicular to H_0)

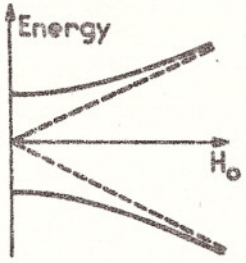
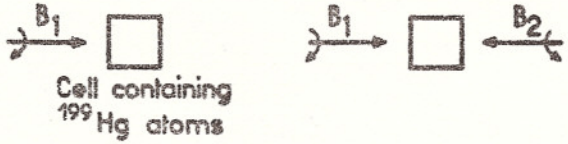


Fig. 7

C. Experimental observations on ^{199}Hg. (1) *General principles:* With a first light beam B_1 we do an ordinary optical pumping experiment† to measure with great precision the Zeeman splitting ω_g between the two sublevels of the ground state of ^{199}Hg (Fig. 8a)

We then add a second light beam B_2 (Fig. 8b) and remeasure, with the help of B_1, the energy difference between the two sublevels of the ground

state. We determine in this way the perturbation of the ground state due to the B_2 irradiation. The characteristics of the two beams are completely different. B_1 is a "good" pumping beam, containing many resonant photons. B_2 is a "good" shifting beam, with mostly non resonant photons.



(a) Fig. 8 (b)

We shall consider two extreme cases: the high field case where $\mathscr{H}_L \ll \mathscr{H}_z$, and the low field case where $\mathscr{H}_L \gg \mathscr{H}_z$.

(2) *High field case* $\mathscr{H}_L \ll \mathscr{H}_z$

a) First experiment. The pumping beam B_1, circularly polarized, comes from a lamp filled with the isotope ^{204}Hg whose line coincides with the k_0 component $F = \frac{1}{2}$ of ^{199}Hg. The steady magnetic field H_0 is parallel to B_1; the radiofrequency field $H_1 \cos \omega t$ is perpendicular to H_0 (Fig. 9). We measure ω_φ by the usual magnetic resonance technique.

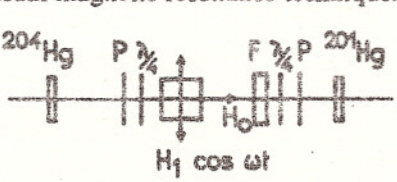


Fig. 9

The second beam B_2 is produced by a lamp filled with the isotope ^{201}Hg whose line center wave number $\bar{k}$ is longer than k_0 by approximately a Doppler width which, according to experimental†† and theoretical (Fig. 5) results, corresponds approximately to the maximum value for $\Delta E'$. A filter F filled with ^{199}Hg is placed before the cell and suppresses all the resonant photons from the B_2 light:

As $\bar{k} - k_0 > 0$, $\Delta E' > 0$: the levels are shifted towards the higher energies.

From the polarization scheme (Fig. 10a) and the expression of the matrix elements of A given in § I-B-2, it appears that: if the polarization of B_2 is σ^+,

† For a detailed description of the optical pumping of the odd isotopes of mercury see B. Cagnac thesis (ref.[1])

†† See later, fig. 12.

the only non zero matrix element of A is $\langle -|A|- \rangle$; thus only the state $|-\rangle$ is shifted (Fig. 10b).

If now the polarization of B_2 is σ^-, we have $\langle +|A|+ \rangle \neq 0$ and only the state $|+\rangle$ is shifted (Fig. 10c).

So the sign of the shift must change with the sense of the circular polarization.

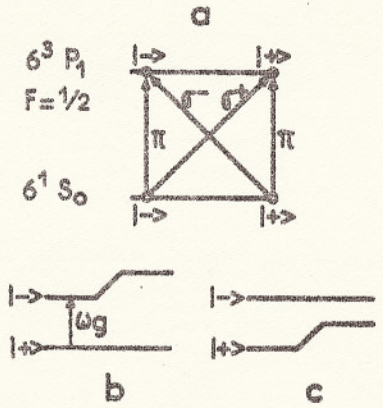


Fig. 10

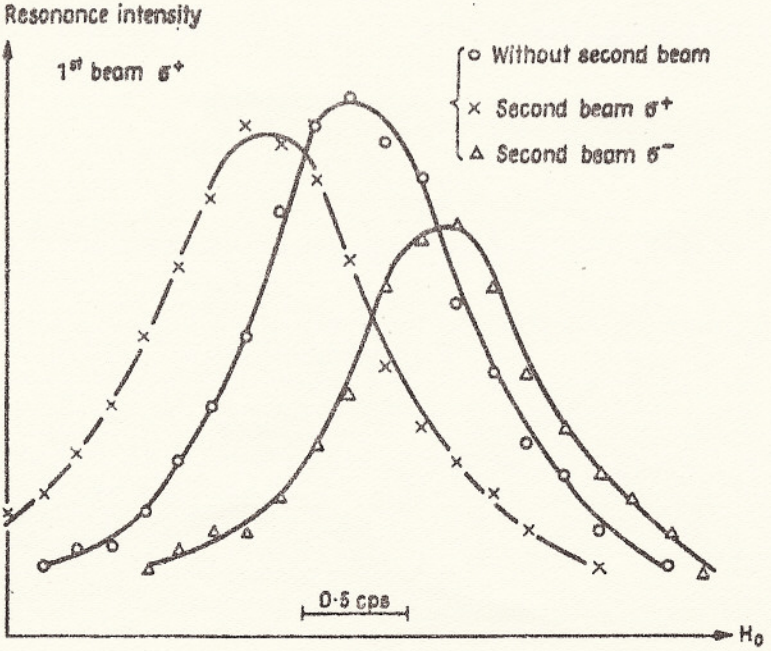


Fig. 11

The experimental results are shown in Fig. 11 and agree with these predictions (as we operate at a fixed frequency and a variable field, an increase of the Zeeman separation corresponds to a shift towards lower field value).

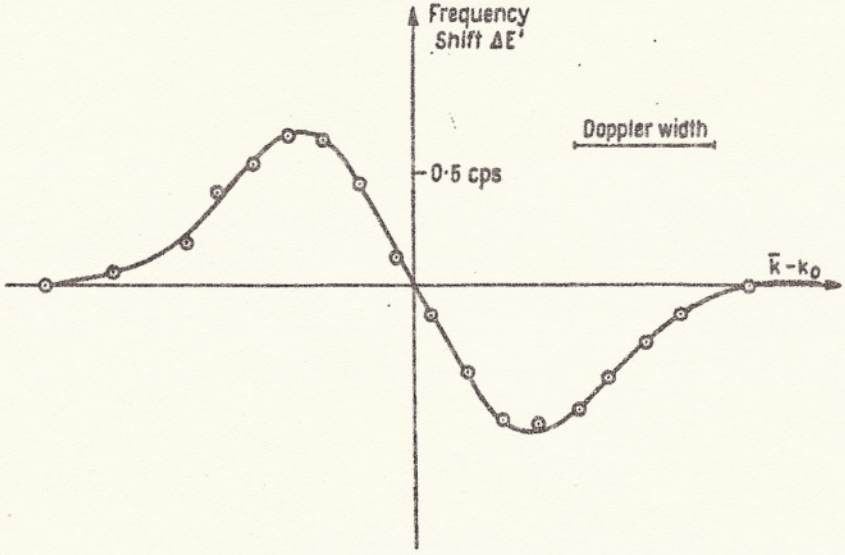


Fig. 12

By magnetic scanning, we have also changed the value of $\bar{k}$ and measured $\Delta E'$ for different $\bar{k} - k_0$. The experimental results are in agreement with the theoretical predictions (Fig. 12)

b) Second experiment. We have improved the first experiment in the following ways:[6]

—The light beam B_1 (Fig. 13) is perpendicular to the magnetic fields H_0 and $H_1 \cos \omega t$ which are now parallel. Using the technique of the transverse

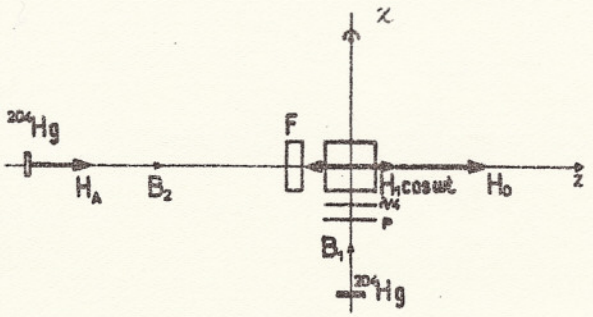


Fig. 13

optical pumping†, we have obtained resonance curves with no RF broadening and whose width is typically of the order of 0.3 hz.

—The second beam B_2, parallel to H_0, is produced by a ^{204}Hg lamp placed in an axial magnetic field H_A. The line emitted is then split into a σ^+ component which has a $\bar{k} > k_0$ and shifts the state $|-\rangle$ towards the higher energies and a σ^- component which has a $\bar{k} < k_0$ and shifts the state $|+\rangle$ towards the lower energies (Fig. 14a and 14b)

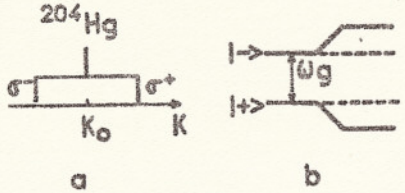


Fig. 14

As the effects of the two components add, there is no need to place, in the B_2 beam's way, a polarizer which absorbs a large part of the incident intensity. We thus get a larger shift $\Delta E'$ and double the effect as is shown in Fig. 14b.

Figure 15 is an example of the experimental results we have obtained.

To avoid any broadening due to real transitions, we still have to use a

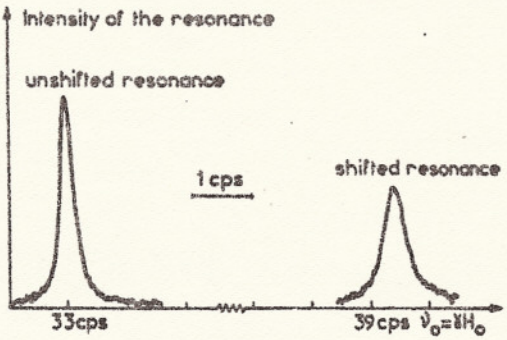


Fig. 15

filter F in the B_2 beam. However, as this filter is not perfect, the shifted curve is slightly broader than the original curve.

The shift obtained is approximately equal to 20 times the line width of the resonance curves.

† in an amplitude modulated field ([7])

(3) *Low field case:* $\mathcal{H}_L \gg \mathcal{H}_z$. As the shift obtained in the previous experiment is much larger than the energy level width, we have been able to study the removal of the Zeeman degeneracy in a zero magnetic field.[8]

The direction of the two beams is the same as that of Fig. 13.

To eliminate any stray magnetic fields we have placed the resonance cell in a triple magnetic shield.

The experiment is performed in the following way: in a zero magnetic field, the magnetic dipoles are first oriented by the beam B_1 in the Ox direction (Fig. 16a)

We then suddenly introduce B_2: the dipoles start to precess around the fictitious magnetic field $\vec{H}_f$ which is associated with B_2 and which is parallel

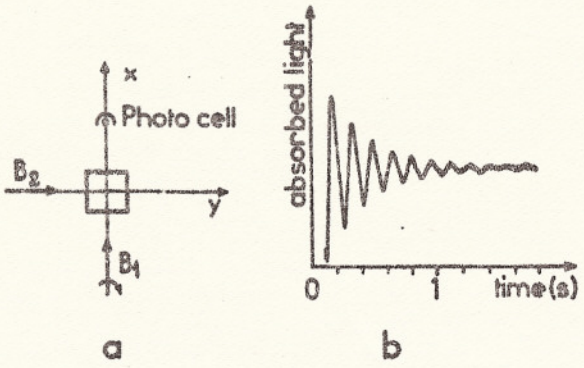


Fig. 16

to Oy; as the shift is much larger than the width of the levels, many oscillations occur during the life time of the ground state. This effect may be observed on the transmitted light along the Ox direction (Fig. 16b).

We have also studied experimentally the Zeeman energy diagram in the presence of the B_2 light for two different cases: $\vec{H}_0$ and $\vec{H}_f$ parallel or perpendicular.

The energy difference ω_0' between the two Zeeman sublevels has been measured

a) by the resonances described in § I-B-2 (transverse optical pumping in an amplitude modulated field)

b) by modulating the polarization of the pumping beam B_1 at the frequency ω[9]; resonances occur when $\omega = \omega_0'$.

For $\vec{H}_0 // \vec{H}_f$, $\omega_0' = \omega_0 + \omega_f$ (ω_f is the Larmor frequency associated with the fictitious field $\vec{H}_f$), we get a displaced Zeeman diagram.

For $\vec{H}_0 \perp \vec{H}_f$, $\omega_0' = \sqrt{\omega^2 + \omega_f^2}$, the two levels do not cross any more. This phenomenon might be compared to the Back-Goudsmit effect, with the light beam B_2 playing the role of the hyperfine structure.

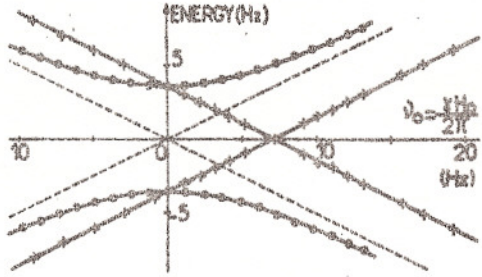


Fig. 17

$$\begin{cases} \times & \vec{H}_0 \parallel \vec{H}_f \\ \circ & \vec{H}_0 \perp \vec{H}_f \end{cases}$$

II. Crossings and Anticrossings of Atomic Energy Levels

Introduction

We will show in this lecture that important variations of the resonance radiation scattered by an atom occur in the neighborhood of what is called a "crossing" or an "anticrossing" point.

We shall first give a definition of a crossing or anticrossing point. We shall then determine the scattering cross section for resonance radiation at these points.

Those effects are very important in atomic spectroscopy. They allow a simple and precise determination of atomic structures and radiative lifetimes. We will also need the concepts introduced here for the interpretation of the effects described in the two last lectures.

A. Crossing

(1) *Definition:* Let us consider an atom which has three levels: a ground state $|g\rangle$ and two excited states $|e_1\rangle$ and $|e_2\rangle$, both having the same lifetime. Let $\mathcal{H}_0$ be the atomic hamiltonian. We have:

$$\mathcal{H}_0 |g\rangle = E_g |g\rangle \quad \mathcal{H}_0 |e_i\rangle = E_{e_i} |e_i\rangle \quad i = 1, 2$$

We shall call $k_i = E_{e_i} - E_g$.

Let us assume that, as we plot the energy of these states as functions of the magnetic field H_0 (Fig. 1), the two excited levels cross for $H_0 = H_c$; at this crossing point, these two excited states have the same energy k_c.

We define the principal polarizations $\vec{\varepsilon}_1$ and $\vec{\varepsilon}_2$ in the following way: absorbing a photon of polarization $\vec{\varepsilon}_1(\vec{\varepsilon}_2)$, the atom can only jump from the ground state to the excited state $|e_1\rangle(|e_2\rangle)$. We have, therefore, the following relations:

$$\langle e_1 |\vec{\varepsilon}_1 \vec{D}| g\rangle \neq 0 \qquad \langle e_2 |\vec{\varepsilon}_1 \vec{D}| g\rangle = 0$$

$$\langle e_1 |\vec{\varepsilon}_2 \vec{D}| g\rangle = 0 \qquad \langle e_2 |\vec{\varepsilon}_2 \vec{D}| g\rangle \neq 0$$

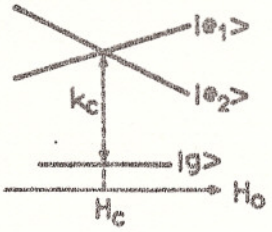


Fig. 1

Any linear superposition $\vec{\varepsilon}$ of $\vec{\varepsilon}_1$ and $\vec{\varepsilon}_2$ is called a "coherent polarization":

$$\vec{\varepsilon} = \lambda_1 \vec{\varepsilon}_1 + \lambda_2 \vec{\varepsilon}_2$$

Absorbing a photon of polarization $\vec{\varepsilon}$, the atom, starting from the ground state, can go to $|e_1\rangle$ and to $|e_2\rangle$. *Both* matrix elements $\langle e_1| \vec{\varepsilon} \cdot \vec{D} |g\rangle$ and $\langle e_2| \vec{\varepsilon} \cdot \vec{D} |g\rangle$ are different from zero.

Fig. 2

(2) *Resonant scattering amplitude.* We deal now with the following problem: sending a photon $(\vec{k}, \vec{\varepsilon})$ on the atom, we look for the scattering amplitude for reemission of a photon $(\vec{k}', \vec{\varepsilon}')$ (Fig. 2)

The two physical processes which may occur are drawn on Fig. 3:

We suppose that we are near resonance, so that k and k' are close to k_1 and k_2. In this case, the second process is negligible (antiresonant) and the

scattering amplitude S_j corresponding to the first process may be written as:[4]

$$S_j = C'^{e} \delta(k - k') \frac{\langle g\vec{k}'\vec{\varepsilon}' | V | e_j \rangle \langle e_j | V | g\vec{k}\vec{\varepsilon} \rangle}{k - k_j + i\frac{\Gamma}{2}} \qquad \text{(II.1)}$$

The δ function is nothing but the energy conservation requirement.

The resonant character of the scattering appears in the denominator; the angular dependence of the scattering is contained in the numerator, whose explicit form is $|A_k|^2 \langle g | \vec{\varepsilon}' \vec{D} | e_j \rangle \langle e_j | \vec{\varepsilon} \cdot \vec{D} | g \rangle$.

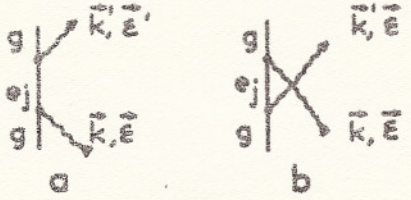


Fig. 3

There are two possible intermediate states for the scattering: $|e_1\rangle$ and $|e_2\rangle$ (Fig. 4); each path corresponds to a different scattering amplitude S_1 or S_2

The total scattering amplitude S is the sum of S_1 and S_2 *which are both different from zero if and only if* $\vec{\varepsilon}$ *and* $\vec{\varepsilon}'$ *are coherent polarizations.* We have then

$$S = C'^{e} \delta(k - k') [R_1 + R_2] \qquad \text{(II.2)}$$

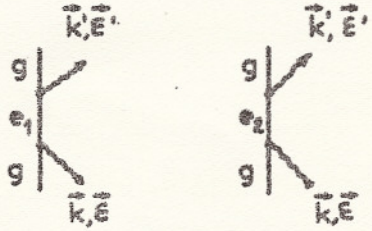


Fig. 4

with

$$R_i = \frac{B_i}{k - k_i + i\frac{\Gamma}{2}} \qquad \text{(II.3)}$$

where

$$B_i = |A_k|^2 \langle g | \vec{\varepsilon}' \vec{D} | e_i \rangle \langle e_i | \vec{\varepsilon} \vec{D} | g \rangle$$

(3) *Resonant scattering cross section* σ. σ is proportional to $|S|^2$. Thus from § II-A-2 we have:

$$\sigma = C^{te} [|R_1|^2 + |R_2|^2 + 2\mathrm{Re}\,(R_1 R_2^*)] \qquad \text{(II.4)}$$

It will appear later that the interference term $\mathrm{Re}\,(R_1 R_2^*)$ is responsible for the resonant variation of σ at the crossing point.

From (II.3), one sees immediately that $R_1(R_2)$ is important only for $|k - k_1| \lesssim \Gamma (|k - k_2| \lesssim \Gamma)$. Therefore the interference term which depends on $R_1 R_2^*$ is important only for $|k - k_1| \lesssim \Gamma$ *and* $|k - k_2| \lesssim \Gamma$, i.e. for $|k_1 - k_2| \lesssim \Gamma$. The effects associated with this term will appear only at the crossing point.

So far, we have considered monochromatic excitation. In a real experiment, we have a broad line excitation $u(k)$ plotted on Fig. 5:

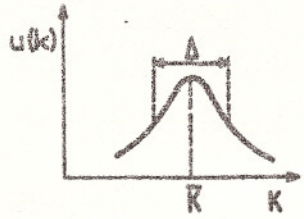


Fig. 5

We assume that

$$\Delta \gg \Gamma \qquad \text{(II.5)}$$

and k_1, k_2, k_c are very close to $\bar{k}$, so that

$$u(k_1) \simeq u(k_2) \simeq u(k_c) \simeq u(\bar{k}) \qquad \text{(II.6)}$$

So far, we have computed the cross section $\sigma(k)$ for a single incident photon k (II.4). As there is no phase relation between photons of different k, we must average the *cross section* (and not the scattering amplitude) over the wave number of the incident photons.

The cross section $\bar{\sigma}$ that we measure in a real experiment is thus:

$$\bar{\sigma} = \int u(k)\, \sigma(k)\, dk$$

Taking into account the conditions (II.5) and (II.6), we get

$$\int |R_f(k)|^2\, u(k)\, dk \simeq 2\pi \frac{|B_f|^2}{\Gamma} u(\bar{k})$$

and

$$2\int \mathrm{Re}\,(R_1(k)\,R_3^*(k))\,u(k)\,dk = 2\mathrm{Re}\,\frac{B_1B_2^*}{k_1-k_2-i\Gamma}\times$$

$$\times\left[\int\frac{u(k)\,dk}{k-k_1+i\dfrac{\Gamma}{2}}-\int\frac{u(k)\,dk}{k-k_2-i\dfrac{\Gamma}{2}}\right]$$

$$\simeq 4\pi\mathrm{Re}\,\frac{B_1B_2^*}{\Gamma+i(k_1-k_2)}\,u(\bar{k})$$

We finally have

$$\bar{\sigma} = C''\left[\frac{|B_1|^2}{\Gamma}+\frac{|B_2|^2}{\Gamma}+2\mathrm{Re}\,\frac{B_1B_2^*}{\Gamma+i(k_1-k_2)}\right] \qquad \text{(II.7)}$$

The interference term gives rise to a Lorentzian resonance (absorption or dispersion shaped according to $B_1B_2^*$, i.e. to $\vec{e}$ and $\vec{e}'$) in the scattering cross section.

If we plot the scattered light in function of H_0, we get, in the case of an absorption shape, the curve shown on Fig. 6.

The width of the resonance ΔH is determined by the condition $|k_1 - k_2| = \Gamma$; it depends only on the natural width Γ and on the slopes of the crossing levels.

It must also be emphasized that, apart from some very small relativistic corrections, the effects we have described do not depend on the velocity of the atom. So the resonance of Fig. 6 is not Doppler broadened.

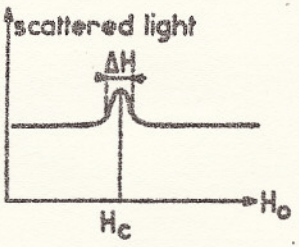


Fig. 6

Let us finally give some well known examples of crossing points.

—Zero field Zeeman crossing: the corresponding resonance is the Hanle effect or the magnetic depolarization phenomenon, discovered in 1924.[10] It allows measurement of either Γ or the Landé factor g of the levels.

—Hyperfine or fine structure crossings: they have been discovered by Franken.[11] From the positions of the crossing points, one can deduce the zero field atomic structure.

B. Anticrossing

(1) *Definition:* We use the same notations as for the crossing case. We assume now that, in addition to the atomic hamiltonian $\mathcal{H}_0$, we have a perturbation h which couples the two excited states $|e_1\rangle$ and $|e_2\rangle$. The only non zero matrix element of h is

$$\delta = \langle e_1 |h| e_2 \rangle$$

The new energy levels $|\bar{e}\rangle$ and $|\bar{e}'\rangle$ of $\mathcal{H}_0 + h$ do not cross each other anymore. The minimum distance between those two levels occurs for $H_0 = H_c$ and is equal to 2δ (Fig. 7). Such a situation is called an anticrossing.

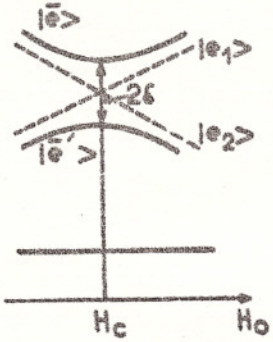


Fig. 7

Let us take now an example where anticrossings can occur.

We consider an atom which has a nuclear spin $\vec{I}$ and an electronic angular momentum $\vec{J}$ in the excited state. In a magnetic field H_0 parallel to Oz, the total hamiltonian is:

$$\mathcal{H} = \omega_e J_z + \omega_n I_z + A\vec{I} \cdot \vec{J}$$

where ω_e and ω_n are the Larmor frequencies of $\vec{J}$ and $\vec{I}$ around H_0, and A the hyperfine coupling constant.

We may write $\mathcal{H}$ as the sum of two hamiltonians

$$\mathcal{H} = \mathcal{H}_0 + h$$

with

$$\begin{cases} \mathcal{H}_0 = \omega_e J_z + \omega_n I_z + A I_z J_z \\ h = \dfrac{A}{2}(I_+ J_- + I_- J_+) \end{cases}$$

as usual

$$I_\pm = I_x \pm iI_y$$

$$J_\pm = J_x \pm iJ_y$$

$|e_1\rangle$ and $|e_2\rangle$ are two eigenstates of $\mathscr{H}_0$, labelled $|m_I m_J\rangle$ with the same value of $m_F = m_I + m_J$: h has then a matrix element between these two states. If for some value of $\mathscr{H}_0$ the two states $|e_1\rangle$ and $|e_2\rangle$ have the same energy, we have an anticrossing situation.

(2) *Problem:* The problem we want to solve is the follwing. Suppose that at time $t = 0$ the atom is excited in the state $|e_1\rangle$ (this may be achieved by a pulse of light of polarization $\vec{\varepsilon}_1$. As the coupling between the atom and the radiation field is purely electronic, one prepares in this way eigenstates of $\mathscr{H}_0$). We ask now for the probability $P(t)$ that the atom is in the state $|e_2\rangle$ at time t (by looking for example at the light polarization $\vec{\varepsilon}_2$ emitted at that time). This physical process is diagrammatically represented as:

If we take into account the life time $1/\Gamma$ of the excited state, we get the probability $\bar{P}$ of having a photon $\vec{\varepsilon}_2$ reemitted after excitation by a photon $\vec{\varepsilon}_1$:

$$\bar{P} = \Gamma \int P(t)\, e^{-\Gamma t}\, dt \qquad \text{(II.8)}$$

$\bar{P}$ is actually the quantity which is measured in an anticrossing experiment.

(3) *Calculation of $P(t)$ and $\bar{P}$ in a simple case.* Let us consider first an atom which has no other excited states then $|e_1\rangle$ and $|e_2\rangle$; h is a non diagonal hamiltonian:

$$\begin{cases} \langle e_i |h| e_i \rangle = 0 \quad \text{for} \quad i = 1, 2 \\ \langle e_1 |h| e_2 \rangle = h_{12} = \delta \end{cases}$$

$P(t)$ may be easily determined by an elementary quantum mechanical calculation. Nevertheless, we shall use the more sophisticated resolvant formalism so as to be able to generalize the results to more complicated cases.

Let us call G and G_0 the resolvant of $\mathscr{H} = \mathscr{H}_0 + h$ and $\mathscr{H}_0$:

$$G = \frac{1}{z - \mathscr{H}_0 - h} \quad \text{and} \quad G_0 = \frac{1}{z - \mathscr{H}_0}$$

As we have shown in a previous chapter (eq. I.4):

$$G = G_0 + G_0 h G$$

By iteration, we derive:

$$G = G_0 + G_0 h G_0 + G_0 h G_0 h G$$

We have to compute the matrix element $\langle e_2 | G | e_1 \rangle = G_{21}$.
Assuming that $\mathcal{H}_0 | e_i \rangle = E_i | e_i \rangle$ $(i = 1, 2)$, we get:

$$G_{21} = \frac{1}{z - E_2} h_{21} \frac{1}{z - E_1} + \frac{1}{z - E_2} h_{21} \frac{1}{z - E_1} h_{12} G_{21}$$

or

$$[(z - E_2)(z - E_1) - |\delta|^2] G_{21} = h_{21}$$

which comes to:

$$G_{21} = \frac{\delta^*}{(z - E_2)(z - E_1) - |\delta|^2} \tag{II.9}$$

G_{21} has two poles, z_+ and z_-, given by:

$$z_\pm = \frac{E_1 + E_2}{2} \pm \sqrt{|\delta|^2 + \left(\frac{E_2 - E_1}{2}\right)^2}$$

We may now compute $U_{21} = \langle e_2 | U(t) | e_1 \rangle$ (see relation I.2):

$$U_{21} = \frac{1}{2\pi i} \int_c G_{21}(z)\, e^{-izt}\, dz$$

$$= \delta^* \left[\frac{e^{-iz_+ t}}{z_+ - z_-} + \frac{e^{-iz_- t}}{z_- - z_+} \right]$$

One gets for $P(t)$ the Breit-Rabi formula:

$$P(t) = |U_{21}(t)|^2 = \frac{|\delta|^2}{|\delta|^2 + \left[\frac{E_2 - E_1}{2}\right]^2} \sin^2 \sqrt{|\delta|^2 + \left(\frac{E_2 - E_1}{2}\right)^2}\, t \tag{II.10}$$

If the excited levels have infinite life times, the probability that the atom passes from the state $|e_1\rangle$ to the state $|e_2\rangle$ is periodical in time. It has a maximum amplitude at the anticrossing point where $E_2 = E_1$.

When we take into account the finite life times of the excited states, we get from (II.8):

$$P = \frac{2 |\delta|^2}{\Gamma^2 + 4 |\delta|^2 + (E_2 - E_1)^2} \tag{II.11}$$

—$\bar{P}$ is resonant at the anticrossing point.

—When $|\delta| \ll \Gamma$, the intensity of the resonance is proportional to $|h|^2$. For $|\delta| \gg \Gamma$, the intensity is constant: we have a saturation of the resonance.

—The width of the resonance is $\sqrt{\Gamma^2 + 4|\delta|^2}$. Contrary to the resonance which occurs at a crossing point, this resonance has a width which depends not only on Γ but also on $|\delta|$. For $|\delta| \gg \Gamma$, the width is proportional to $|\delta|$.

(4) *Calculation of $P(t)$ and $\bar{P}$ in the general case.* We now consider excited states other than $|e_1\rangle$ and $|e_2\rangle$: Let us call them $|e_\alpha\rangle$. We assume that h couples $|e_1\rangle$ and $|e_2\rangle$ to the $|e_\alpha\rangle$, but not necessarily $|e_1\rangle$ to $|e_2\rangle$: we may have $\langle e_1| h |e_2\rangle = 0$; nevertheless $|e_1\rangle$ and $|e_2\rangle$ are still coupled by higher order terms; for instance: $\langle e_2| h |e_\alpha\rangle \langle e_\alpha| h |e_1\rangle \neq 0$.

We shall use the results of § I.A and consider a subspace ε subtended by the two vectors $|e_1\rangle$ and $|e_2\rangle$. We have to compute $\bar{G}(z) = PG(z)P$ with $P = |e_1\rangle\langle e_1| + |e_2\rangle\langle e_2|$ and $Q = 1 - P$.

We get (see I.7):

$$\bar{G}(z) = \frac{1}{z - \overline{\mathscr{H}}_0 - \bar{R}(z)}$$

with

$$\bar{R}(z) = h + Ph\frac{Q}{z - \mathscr{H}_0}hP + \cdots \qquad \text{(II.12)}$$

$\bar{G}(z)$ is the inverse of the two by two matrix $z - \overline{\mathscr{H}}_0 - \bar{R}(z)$:

$$\begin{pmatrix} z - E_1 - \bar{R}_{11} & -\bar{R}_{12} \\ -\bar{R}_{21} & z - E_2 - \bar{R}_{22} \end{pmatrix}$$

It is easy to invert such a matrix, and one gets for $\bar{G}_{21}(z)$:

$$\bar{G}_{21}(z) = \frac{\bar{R}_{21}(z)}{[z - E_1 - \bar{R}_{11}(z)][z - E_2 - \bar{R}_{22}(z)] - |\bar{R}_{12}(z)|^2} \qquad \text{(II.13)}$$

So far, the formula is rigorous.

We shall now make the same approximation as in § A.I and II and replace in $\bar{R}(z)$ z by E_c where E_c is the common value of E_1 and E_2 at the crossing point. This approximation is justified if the energies E_α of the other excited states $|e_\alpha\rangle$ are far enough from E_1 and E_2:

$$|E_{1,2} - E_\alpha| \gg |\langle e_{1,2}| h |e_\alpha\rangle|$$

We then get:

$$G_{21}(z) = \frac{\bar{R}_{21}(E_c)}{[z - E_1 - \bar{R}_{11}(E_c)][z - E_2 - \bar{R}_{22}(E_c)] - |\bar{R}_{21}(E_c)|^2} \qquad \text{(II.14)}$$

Let us compare this formula to the result of the previous section: we see that if we replace, in equ. (II.9), E_i by $E_i + \bar{R}_{ii}(E_c)$ ($i = 1$ or 2) and h_{21} by $\bar{R}_{21}(E_c)$, we get the same equation as (II.14).

. Using now equation (II.11), we get directly the probability $\bar{P}$ we are looking for:

$$\bar{P} = \frac{2\,|\bar{R}_{21}|^2}{\Gamma^2 + 4\,|\bar{R}_{21}|^2 + [E_2 + \bar{R}_{22} - E_1 - \bar{R}_{11}]^2} \qquad \text{(II.15)}$$

The interpretation of this formula is the following:

—Because of the coupling with the states $|e_\alpha\rangle$, $|e_1\rangle$ and $|e_2\rangle$ are shifted by the quantity $\bar{R}_{11}$ and $\bar{R}_{22}$. This shift is at least of the second order:

$$\bar{R}_{ii} = \sum_\alpha \frac{|h_{\alpha i}|^2}{E_c - E_\alpha} \qquad i = 1, 2 \quad \alpha \neq 1, 2$$

which is the well-known second order perturbation formula. The crossing point H_c is shifted to H_c' (Fig. 8).

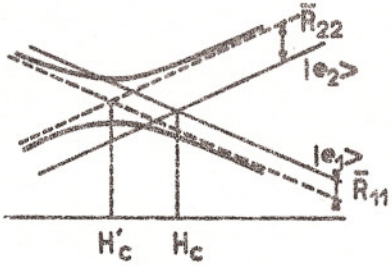


Fig. 8

—The coupling between $|e_1\rangle$ and $|e_2\rangle$ is more complicated. From eq. (II.12), we see that even if $h_{21} = 0$, the two levels are coupled, for instance to second order by $\bar{R}_{12} = \sum_\alpha \frac{h_{1\alpha}\, h_{\alpha 2}}{E_c - E_\alpha}$; the crossing becomes an anti-crossing which is centered at H_c'. Such a situation where $h_{12} = 0$ but $R_{12} \neq 0$ is called a higher order anticrossing.

III. Interaction with RF Photons

We shall report in the first part (§ A) some experimental results concerning the interaction between atoms and an RF field; we shall explain these results in the second part (§ B) by using the concepts of crossing and anticrossing.

A. Description of experimental facts

We shall speak about experiments performed in the ground state of ^{199}Hg ($I = \frac{1}{2}$) which has only two Zeeman sublevels $|+\rangle$ and $|-\rangle$.

1) *Multiple quantum transitions.*† *Experimental set-up:* The cell containing the ^{199}Hg atoms is placed in a magnetic field H_0, parallel to Oz (Fig. 1a).

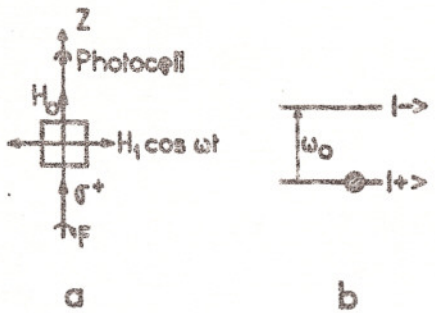


Fig. 1

The energy difference between the two sublevels is $\omega_0 = \gamma H_0$ ($\hbar = 1$); γ is the gyromagnetic ratio of the ground state ($\gamma < 0$). The atoms are optically pumped by a σ^+ polarized light beam F parallel to H_0: most of the atoms are then in the state $|+\rangle$ (Fig. 1b).

Any change in the population of the two states may be detected by measuring the intensity I of the transmitted pumping light.

We apply a RF field $H_1 \cos \omega t$ perpendicular to H_0. ω being fixed, we observe, as we vary ω_0, several resonant variations of I. Transitions between $|+\rangle$ and $|-\rangle$ are induced by the RF field.

Description of the resonances. We observe an *odd spectrum* of resonances: they occur for $\omega_0 = (2n + 1)\,\omega$ (where n is an integer). They are broadened and shifted as we increase the amplitude of H_1 (Fig. 2).

Qualitative interpretation. The linear RF field can be decomposed into two rotating fields. Therefore the quantized field contains photons whose polarization is either σ^+ or σ^- with respect to Oz. In a real RF transition, the total angular momentum and the energy have to be conserved. σ^+ and σ^- RF photons have an angular momentum $+1$ or -1, and an energy

† The first observation of multiple quantum transitions on optically pumped atoms has been performed on alkali atoms[12]. Winter's thesis contains the first theoretical explanation of this penomenon. We will use her- a different theoretical approach which also applies to the more recently discovered effects described in § III·A-2 and § III A-3.

ω ($\hbar = 1$). Transitions between the atomic states $|+\rangle$ and $|-\rangle$ may thus occur in the following cases:

$\omega_0 = \omega$ and the atom absorbs one photon σ^- (Fig. 3a).

$\omega_0 = 3\omega$ and the atom absorbs 3 photons, one σ^+ and two σ^- (Fig. 3b) more generally, one needs an odd number of RF photons to satisfy both angular momentum and energy conservations:

$\omega_0 = (2n + 1)\,\omega$ and the atom absorbs n photons σ^+ and $(n + 1)$ photons σ^-.

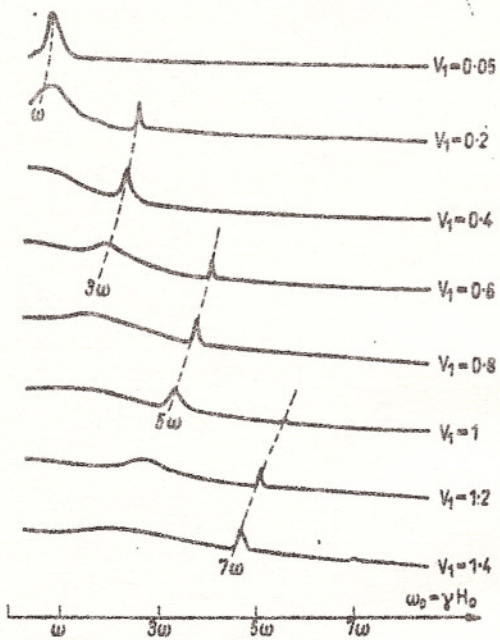


Fig. 2 (each curve corresponds to a different value of H_1 measured by the parameter V_1 (voltage at the RF coils))

As one increases the intensity of the RF field, more transitions between $|-\rangle$ and $|+\rangle$ may occur; the lifetimes of those states are therefore reduced and the resonances broadened.

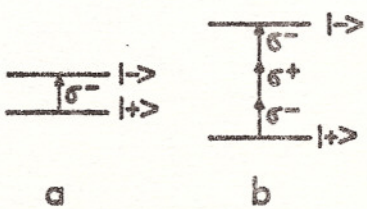


Fig. 3

Bloch-Siegert[13] type shifts also occur: as we increase H_1, the resonances are shifted towards the low field region.

(2) *Transverse optical pumping. Haroche's resonances* [14]. The *experimental set-up* is very similar to that of the previous section (§ III.A.1) except for the direction of F which is now perpendicular to H_0 (Fig. 4). This is a case of transverse optical pumping. Let us recall briefly some of its features[1]:

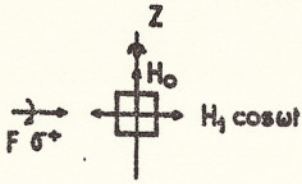


Fig. 4

Transverse optical pumping ($H_1 = 0$). No population difference is produced between the two states $|+\rangle$ and $|-\rangle$; in other words, no longitudinal magnetization is introduced by the pumping.

If $H_0 = 0$, F orients the vapor's magnetic dipoles $\vec{M}$ in its own direction (Fig. 5a). In mathetmatical terms, the density matrix which describes the atoms in the ground state, has non-zero off diagonal elements (in the Oz representation).

If $H_0 \neq 0$, as soon as the dipoles are oriented, they start to precess around H_0. They are damped at a rate $1/T$ because of the relaxation (Fig. 5b. H_0 is perpendicular to the figure). The resulting orientation at time $t = 0$ is the

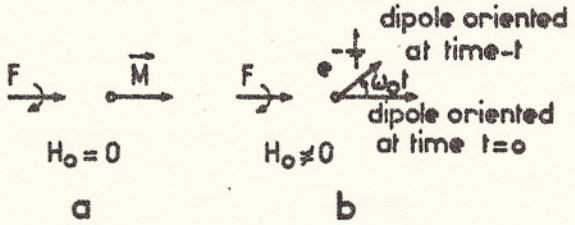


Fig. 5

vectorial sum of all the dipoles created at time $-t$ (t goes from 0 to $+\infty$). They have an amplitude proportional to $e^{-t/T}$ and make an angle $\omega_0 t$ with their initial direction.

Let us make this summation for two extreme cases (Fig. 6a and 6b):

Hence the transverse pumping creates a magnetization in the vapor only for small fields. This is the Hanle effect[10], or the zero field level crossing.

The experimental facts we shall describe now occur for $\omega_0 \gg 1/T$, so that there is no orientation (longitudinal or transverse) introduced by the beam F. We apply a RF field $H_1 \cos \omega t$ parallel to F (Fig. 4). Keeping ω fixed, we vary ω_0 and look at the absorbed light of the pumping beam.

Description of Haroche's resonances. No resonance appears for $\omega_0 = (2n + 1)\omega$. Multiple quantum transitions actually occur, but they are no longer detectable, because the two levels $|-\rangle$ and $|+\rangle$ are equally populated.

— New resonances appear for $\omega_0 = 2n\omega$. They form an *even spectrum*, and may be detected on the various even harmonics $2p\omega$ of the signal. As

The net resultant is zero

The net resultant is not equal to zero

Fig. 6

the intensity H_1 increases, the resonances *are shifted* but *not broadened* as is shown on the next 2 figures. Each resonance corresponds to a different value of H_1, measured by the parameter V_1:

One can show that the shift and the intensity (at the harmonic 2ω) of the resonance $\omega_0 = 2\omega$ are proportional to H_1^2; all the peaks of the curves of Figure 7 are thus on a straight line.

For $\omega_0 = 4\omega$, the shift of the resonances can be shown to be proportional to H_1^2, the intensity (at the harmonic 4ω) to H_1^4: the peaks of the curves of Figure 8 must therefore be on a parabola (plotted with dotted line).

It is impossible to attribute these resonances to the absorption of an even number of RF photons: although energy may be conserved in such a process, the angular momentum carried by an even number of photons σ^+ or σ^- cannot be equal to $+1$ or -1 which is the condition to make the atom jump from $|-\rangle$ to $|+\rangle$ or vice versa.

Before giving a theoretical explanation of these resonances, let us mention another type of resonance which leads to similar difficulties.

(3) *Transverse optical pumping. Resonance of Geneux, Alexandrov, Polonsky* [7,15].

intensity of the resonance $\omega_0 = 2\omega$, harmonic 2ω

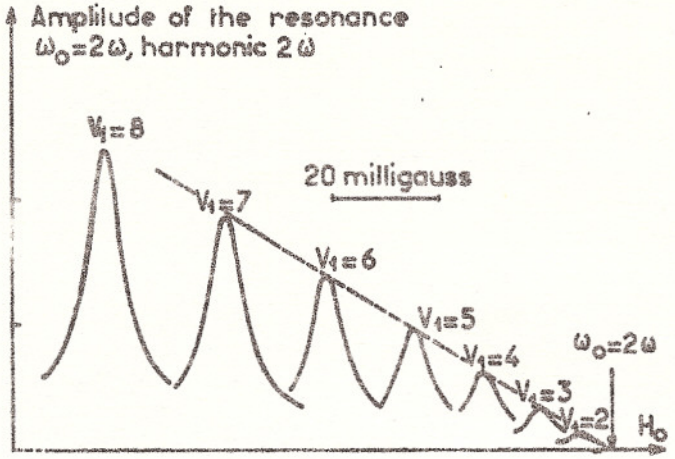


Fig. 7

intensity of the resonance $\omega_0 = 4\omega$, harmonic 4ω

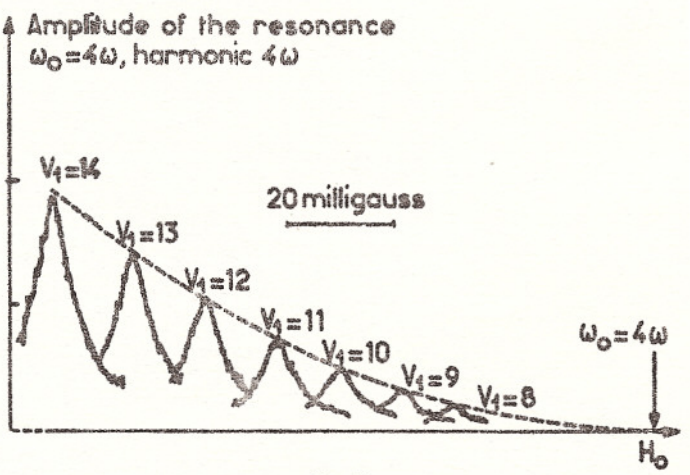


Fig. 8

— *Experimental set-up*: The beam F is still perpendicular to H_0, but $H_1 \cos \omega t$ is now parallel to H_0 (Fig. 9):

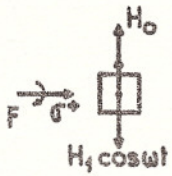


Fig. 9

As the RF field contains only π photons, it is impossible to have any transition from one Zeeman subleved to the other.

Furthermore, the experiment is performed for $\omega_0 \gg \Gamma$: so that there is no transverse pumping.

Nevertheless, as we vary H_0, ω being fixed, and observe the absorption of the pumping light, we get *a full spectrum* of resonances.

— *Description of the resonances:* They occur for $\omega_0 = n\omega$. As the intensity of the RF field is increased, we observe *no shift* and *no broadening* of the resonance curves.

On Figure 10, we have plotted the width of the resonances as a function of $\omega_1/2\pi$ which would be the real value of the resonance's width in an ordinary magnetic resonance:

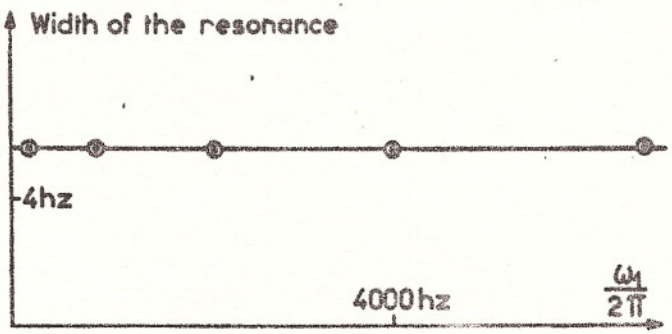


Fig. 10

These resonances may be detected at the various harmonics $p\omega$ of the signal. Their intensity as a function of ω_1/ω has been theoretically predicted and experimentally measured (Fig. 11):

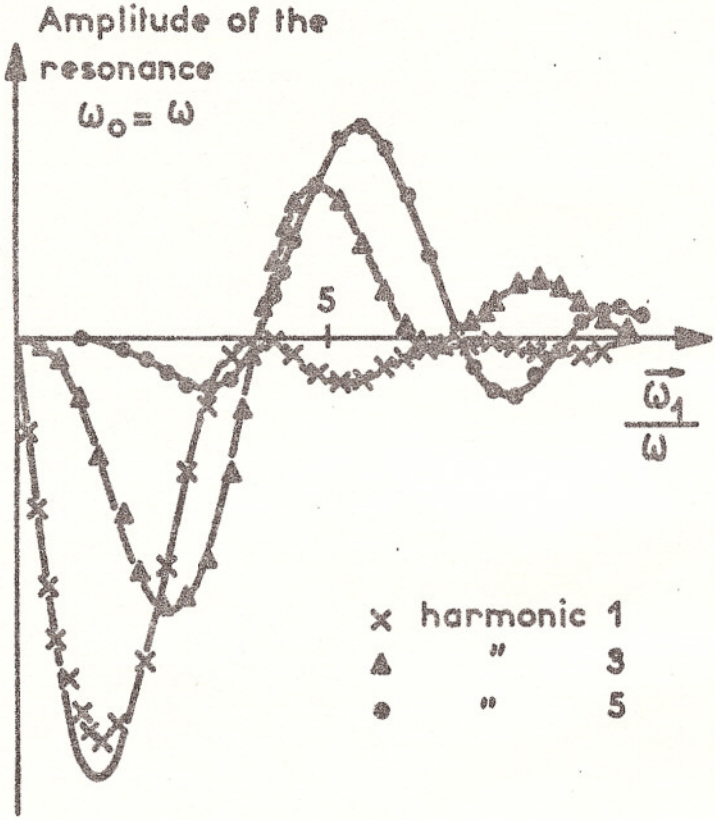


Fig. 11

B. Theoretical interpretation

(1) *Introduction.* We shall give a single theoretical treatment which applies to the three preceding types of resonances.

Let us first define some simplifications we have made in this treatment:

a) although the experimental observations have ben done on a ground state, we shall deal in the theoretical part with an excited state. The physical effects are similar in both cases but the theory is simpler for an excited state because:

1. the sublevels $|+\rangle$ and $|-\rangle$ of an excited state have the same lifetime (which is not true in general for a ground state).
2. there are no complications due to the falling back of atoms after an optical excitation.

b) We also assume that the atoms and the RF field interact only in the excited state. This amounts to assuming that the resonance conditions do not occur simultaneously in both states.

Let us now explain the general idea of this treatment. We shall deal with the total system (S) which includes the atom plus the RF photons. We shall assume that light is not scattered by the "bare" atom, but by (S) which is something like a "dressed atom". Studying the energy levels of (S), we shall find a lot of crossing and anticrossing points. We shall thus interpret the resonances described in the previous section (§ III.A) by using the result of chapter II. It will also be shown that these resonances may be understood in terms of real and virtual multiple quantum transitions.

This study will suggest to us a new effect; the modification of the Landé g-factor of an atom by an RF field:

$$g_{\text{dressed atom}} \neq g_{\text{bare atom}}$$

We shall present a simple calculation of this effect, and some experimental results.

(2) *General form of the hamiltonian of (S): atom + RF field.* It may be written as the sum of two hamiltonians:

$$\mathcal{H} = \mathcal{H}_0 + h$$

$\mathcal{H}_0$ represents the sum of the energies of the atom and of the RF photons, of frequency ω:

$$\mathcal{H}_0 = \omega_0 J_z + \omega a^\dagger a \qquad \text{(III.1)}$$

We assume that the static field H_0 is in the Oz direction, $\omega_0 = \gamma H_0$, γ and J being the gyromagnetic ratio and the angular momentum of the excited state, $a^\dagger$ and a being the creation and annihilation operator of a photon ω.

h is the hamiltonian which couples the atom to the RF field. In the quantized theory of fields, the RF field $H_1 \cos \omega t$ is represented by:

$$H_1 = \frac{\beta}{\sqrt{q}} [a^+ e^{iqx} + a^+ e^{-iqx}]$$

β is a constant, q the wave number of the RF field.

In the dipole approximation, e^{iqx} is approximately equal to 1 and H_1 is proportional to $(a + a^\dagger)$. Thus the full quantized version of the classical interaction hamiltonian $h_{\text{cl.}} = -\gamma H_1 J_t \cos \omega t$ is:

$$h_{\text{qu.}} = \lambda J_t (a + a^\dagger) \qquad \text{(III.2)}$$

where λ is a coupling constant, J_i is the component of angular momentum in the direction of $\vec{H}_1$.

Let us *evaluate the constant* λ:

The quantum state which easily describes the RF field is a coherent state $|\alpha\rangle$[16]. The properties of such a state are well-known: $|\alpha\rangle$ is an eigenstate of a:

$$a|\alpha\rangle = \alpha|\alpha\rangle$$

The parameter α is simply related to the average number $\bar{N}$ of RF photons:

$$\bar{N} = \langle\alpha|a^\dagger a|\alpha\rangle = \alpha^2$$

(we take α real.)

The condition which determines λ is the following:

$$\langle\alpha|h_{qu.}|\alpha\rangle = h_{cl.}$$

From (III.2) it follows:

$$\lambda\langle\alpha|a + a^\dagger|\alpha\rangle = \gamma H_1 = \omega_1$$

or

$$2\lambda\alpha = \omega_1$$

We finally have:

$$\lambda = \frac{\omega_1}{2\sqrt{\bar{N}}} \tag{III.3}$$

Actually, λ may be computed from the first principles. If we consider a coherent state corresponding to a known value of $\bar{N}$, we may deduce from (III.3) the value ω_1 of the classical field described by the coherent state.

We shall study in the following sections two different cases corresponding to $\vec{H}_1$ and $\vec{H}_0$ perpendicular or parallel.

(3) $\vec{H}_1$ *perpendicular to* $\vec{H}_0$:

a) *Hamiltonian:* The geometrical arrangement of $\vec{H}_0$ and $\vec{H}_1$ is plotted in Figure 12:

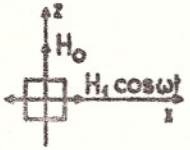


Fig. 12

Using (III.1) and (III.2) we have

$$\mathcal{H} = \underbrace{\omega_0 J_z + \omega a^\dagger a}_{\mathcal{H}_0} + \underbrace{\lambda J_x (a + a^\dagger)}_{h} \tag{III.4}$$

As the RF field interacts only with the excited atoms, the eigenstates of $\mathscr{H}$ in the ground state are very simple:

$$\mathscr{H}\,|\,g,n\,>\,=\,(E_g\,+\,n\omega)\,|\,g,n\,>$$

The problem is more complicated in the excited state. We shall begin to study the eigenstates of $\mathscr{H}_0$.

b) *Energy levels of* $\mathscr{H}_0$

Let us call $|n, \pm\rangle$ the eigenstates of $\mathscr{H}_0$:

$$\mathscr{H}_0|n, \pm\,\rangle\,=\,\left(n\omega\,\pm\,\frac{\omega_0}{2}\right)|n, \pm\rangle$$

We have plotted in Figure 13 the energy levels of $\mathscr{H}_0$ as a function of ω_0 (we suppose here that ω_0 is >0):

This energy diagram shows an infinite number of crossing points: The level $|n, -\rangle$ crosses the level $|n', +\rangle$ for $\omega_0 = (n - n')\,\omega$. We shall call this point an odd (even) crossing if $(n - n')$ is odd (even); it is impoftant to study the effect of h in the neighborhood of those points.

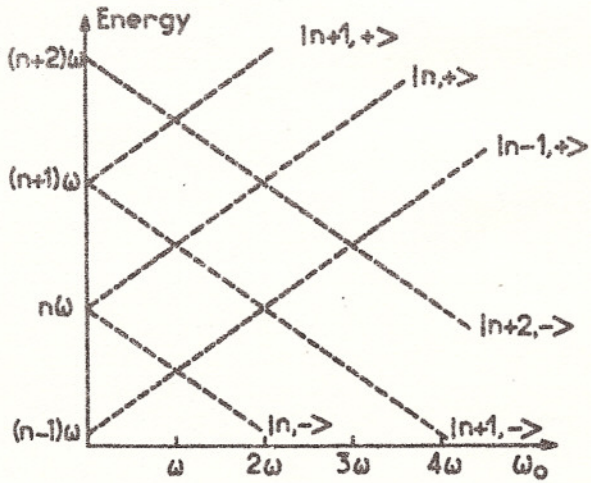


Fig. 13

We consider in this paragraph (B.3) that h is a small perturbation, and more precisely that $\omega_1 \ll \omega_0, \omega$.

c) *Effect of h at an odd crossing*

Let us first study the crossing which occurs for $\omega_0 = \omega$ between the two levels $|n + 1, -\rangle = |e_1\,\rangle$ and $|n, +\rangle = |e_2\rangle$.

Since J_x has a matrix element between $|+\rangle$ and $|-\rangle$ and a and $a\dagger$ have matrix elements between the states $|n\rangle$ and $|n+1\rangle$, the perturbation h couples the two levels $|e_1\rangle$ and $|e_2\rangle$: we are in an *anticrossing* situation. We notice that $|e_1\rangle$ and $|e_2\rangle$ are also coupled to other states, for instance $|n-1, -\rangle$, $|n+2, +\rangle$ etc. Using the results of § II.B.4, we see that under the effect of h, the crossing becomes a shifted anticrossing.

If initially the system is in the state $|n+1, g\rangle$ and if one sends in the atom light with a principal polarization $\vec{\varepsilon}_1$ corresponding to the transition $|g\rangle \leftrightarrow |-\rangle$, one prepares the system in the state $|n+1, -\rangle$ of $\mathscr{H}_0$ (the electric dipole operator and the RF operators commute so that $\Delta n = 0$ in an optical transition). Similarly by looking at the light reemitted with a principal polarization $\vec{\varepsilon}_2$ (corresponding to the transition $|g\rangle \leftrightarrow |+\rangle$) one measures the probability of the transition $|n+1, -\rangle \to |n, +\rangle$ under the effect of h, i.e. the probability that the atom has absorbed one RF quantum, jumping from $|-\rangle$ to $|+\rangle$. To summarize, by sending $\vec{\varepsilon}_1$ light and by looking at $\vec{\varepsilon}_2$ light, one measures the quantity $\bar{P}$ defined in § II.B.4. (see equ. II.15):

$$\bar{P} = \frac{2|\bar{R}_{21}|^2}{\Gamma^2 + 4|\bar{R}_{21}|^2 + [E_2 + \bar{R}_{22} - E_1 - \bar{R}_{11}]^2}$$

Let us calculate the matrix element of R. In the lowest non-zero order, they are:

$$\bar{R}_{21} \simeq h_{21} = \lambda \langle n, + | J_x (a + a^\dagger) | n+1, - \rangle$$

$$= \frac{\lambda}{2}\sqrt{n+1}$$

Using (III.3), we get $\bar{R}_{21} = \frac{\omega_1}{4}$.

Similarly one finds

$$\bar{R}_{11} = \frac{|\langle n+2, + | h | n+1, - \rangle|^2}{E_{(n+1,-)} - E_{(n+2,+)}} = -\frac{\omega_1^2}{32\omega} = -\bar{R}_{22}$$

and finally we get for $\bar{P}$

$$\bar{R} = \frac{\frac{\omega_1^2}{8}}{\Gamma^2 + \frac{\omega_1^2}{4} + \left[\omega_0 - \omega + \frac{\omega_1^2}{16}\right]^2}$$

With the $\vec{\varepsilon}_2$ light the one RF quantum absorption appears as a Lorentzian resonance centered at $\omega_0 = \omega - (\omega_1^2/16\omega)$ with an intensity proportional to ω_1^2 (for $\omega_1 \ll \Gamma$), a width proportional to ω_1, for $\omega_1 \gg \Gamma$.

We shall now study a slightly more complicated case: The crossing between $|e_1\rangle = |n+3, -\rangle$ and $|e_2\rangle = |n, +\rangle$ which occurs for $\omega_0 = 3\omega$.

There is only a third order coupling between $|e_1\rangle$ and $|e_2\rangle$. We get a *higher order anticrossing* situation. The two levels are connected by the real absorption of three RF photons.

Let us calculate the matrix elements of R:

$$R_{12} = \frac{\langle n, + |h| n+1, -\rangle \langle n+1, - |h| n+2, + \rangle \langle n+2, + |h| n+3, -\rangle}{(E_{n,+} - E_{n+1,-})(E_{n,+} - E_{n+2,+})}$$

$$= \frac{\lambda^3}{8} \sqrt{n+3}\sqrt{n+2}\sqrt{n+1}\,\frac{1}{2\omega}\,\frac{1}{-2\omega}$$

Finally

$$R_{12} = -\frac{\omega_1^3}{256\omega^2}$$

In the same way

$$R_{11} = \frac{|\langle n+2, + |h| n+3, -\rangle|^2}{E_{n+3,-} - E_{n+2,+}} + \frac{|\langle n+4, + |h| n+3, -\rangle|^2}{E_{n+3,-} - E_{n+4,+}}$$

$$= -\frac{3\omega_1^2}{64\omega} = -R_{22}$$

So that:

$$P_{21} = \frac{2\left(\frac{1}{256}\right)^2 \frac{\omega_1^6}{\omega^4}}{\Gamma^2 + 4\left(\frac{1}{256}\right)^2 \frac{\omega_1^6}{\omega^4} + \left[\omega_0 - 3\omega + \frac{3}{32}\frac{\omega_1^2}{\omega}\right]^2}$$

We get a resonance centered at $\omega_0 = 3\omega - \frac{3}{32}\frac{\omega_1^2}{\omega}$ whose intensity is proportional to ω_1^6 and whose width varies as ω_1^3 (for $\omega_1 \gg \Gamma$).

More generally, it can be shown that any odd crossing becomes an anticrossing. The corresponding resonance is related to the real absorption of an odd number of RF quanta. This resonance is shifted and broadened as the intensity of the RF field increases. Principal polarizations have to be used for the incoming and outgoing light.

All the experimental features described in § III.A.1 are quantitatively understood.

d) *Effect of h at an even crossing:*

For $\omega_0 = 2\omega$, the two levels $|e_1\rangle = |n, +\rangle$ and $|e_2\rangle = |n+2, -\rangle$ cross each other. When h is applied, $|n, +\rangle$ is coupled to $|n+1, -\rangle$ which is coupled to $|n+2, +\rangle$ etc.

There is therefore no coupling at any order between $|e_1\rangle$ and $|e_2\rangle$. This is a consequence of the conservation of angular momentum. But, as $|e_1\rangle$ and $|e_2\rangle$ are coupled to other states, the crossing point is shifted (see Figure 14)

It is now possible to give an explanation of Haroche's resonances [17]. They correspond to the level crossing resonances of the "dressed" atom. One sees in Figure 14 that the resonances are shifted when H_1 increases; but as we have

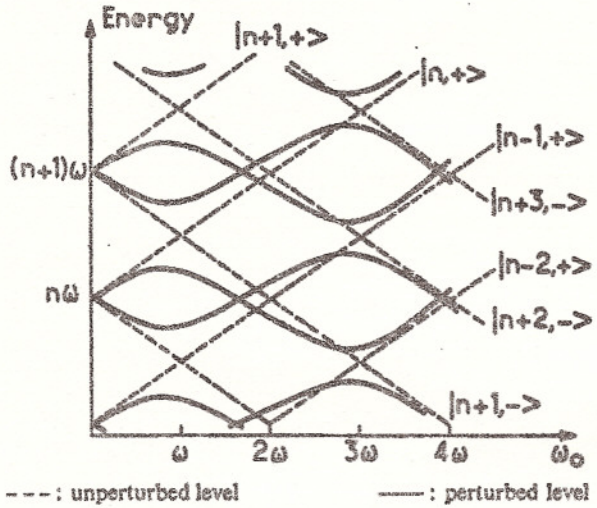


Fig. 14

shown in § II.A, the width of a level crossing resonance depends only on the width and the slopes of the two levels which cross and that explains why one does not observe any RF broadening. One understands also why these resonances appear in transverse optical pumping: a necessary condition for the observation of level crossing resonances is that the polarization of the light must be coherent as we have seen in § II.A; and it can be shown that in transverse optical pumping, this condition is fulfilled.

According to (II.4), the crossing signal is proportional to :

$$\langle \vec{e}_2 \,|\, \vec{e}\,\vec{D} \,|\, gn\rangle \langle gn \,|\, \vec{e}\,\vec{D} \,|\, \vec{e}_1\rangle$$

where $\vec{e}$ is the polarization of the incoming light and $|\vec{e}_1\rangle$, $|\vec{e}_2\rangle$ are the two (perturbed) states which cross.

If instead of $|\vec{e}_1\rangle$ and $|\vec{e}_2\rangle$, we had $|e_1\rangle$ and $|e_2\rangle$, i.e. the two unperturbed states, the matrix elements would be 0 because the matrix element of $\vec{e}\,\vec{D}$ must fulfil the condition $\Delta n = 0$, so that $\langle n+2, -| \vec{e}\,\vec{D} \,|gn\rangle = 0$.

In fact, we must use the perturbation expansion of $|\bar{e}_1\rangle$ and $|\bar{e}_2\rangle$. For example we have

$$|\bar{e}_2\rangle = |n+2,-\rangle + \eta\,|n+1,\ +\rangle + (\eta')^2|\,n,-\rangle + \ldots$$

where η and η' are proportional to the perturbation, that is to say, to ω_1.

In the state $|e_2\rangle$ appears now, to the second order, the state $|n,-\rangle$. We say that the state $|n+2,-\rangle$ is "contaminated" to the second order by $|n,-\rangle$ through "*virtual absorption and reemission of two quanta*". As a consequence of these virtual processes, the matrix elements of formula (III.5) are no longer zero. We have: $\langle\bar{e}_2|\ \vec{\varepsilon}\cdot\vec{D}\,|gn\rangle = (\eta')^2\,\langle n,-|\ \vec{\varepsilon}\,\vec{D}\,|gn\rangle$.

A similar treatment can be applied to other crossing points, where $\omega_0 = 2n\omega$. We find shifted but not broadened resonances, which can be interpreted as being due to virtual absorption and reemission of $2n$ quanta.

(4) $\vec{H}_1$ *parallel to* $\vec{H}_0$. The hamiltonian of the problem is:

$$\mathcal{H} = \underbrace{\omega_0 J_z + \omega a^\dagger a}_{\mathcal{H}_0} + \underbrace{\lambda J_z\,(a + a^\dagger)}_{h}$$

The energy levels of $\mathcal{H}_0$ are plotted in Figure 13. Since J_z has no matrix element between $|+\rangle$ and $|-\rangle$, there is no coupling at any order between two crossing levels $|n,+\rangle$ and $|n',-\rangle$. There is no anticrossing at all in the energy diagram of $\mathcal{H}$.

There is no need, in this particular case, to consider h as a small perturbation. It is possible to find the exact eigenvalues and eigenstates of $\mathcal{H}$[18].

We write $\mathcal{H}$ in the following way:

$$\mathcal{H} = \frac{\varepsilon}{2}\,\omega_0 + V_\varepsilon \qquad \varepsilon = + \text{ or } -$$

with

$$V_\varepsilon = \omega a^\dagger a + \frac{\varepsilon\lambda}{2}\,(a + a^\dagger)$$

or

$$V_\varepsilon = \omega\left(a^\dagger + \frac{\varepsilon\lambda}{2\omega}\right)\left(a + \frac{\varepsilon\lambda}{2\omega}\right) - \frac{\lambda^2}{4\omega} \tag{III.6}$$

Using the property of the displacement operator[12] $D(\varrho)$

$$\begin{cases} D\,a\,D^\dagger = a - \varrho \\ D\,a^\dagger\,D^\dagger = a^\dagger - \varrho \end{cases}$$

We find:

$$V_\varepsilon = D\left(-\frac{\varepsilon\lambda}{2\omega}\right)\left[\omega a^\dagger a - \frac{\lambda^2}{4\omega}\right]D^\dagger\left(-\frac{\varepsilon\lambda}{2\omega}\right)$$

We then easily get the eigenstates $|\bar{n}_\varepsilon\rangle$ of V_ε:

$$|\bar{n}_\varepsilon\rangle = D\left(-\frac{\varepsilon\lambda}{2\omega}\right)|n\rangle \tag{III.7}$$

The eigenstates and eigenvalues of $\mathscr{H}$ are:

$$\mathscr{H}\,|\varepsilon\rangle|\bar{n}_\varepsilon\rangle = \left(\frac{e}{2}\,\omega_0 + n\omega - \frac{\lambda^2}{4\omega}\right)|\varepsilon\rangle\,|\bar{n}_\varepsilon\rangle \tag{III.8}$$

Each energy level of $\mathscr{H}_0$ is shifted by the same quantity $-\dfrac{\lambda^2}{4\omega}$ so that all the crossing points of the energy diagram of $\mathscr{H}$ remain at $\omega_0 = n\omega$.

We understand now the main features of the resonances described in § III.A.(3). We observe in transverse optical pumping a full spectrum of resonances which are not shifted and not broadened by the RF field.

A useful identity may be derived for very large n[18]:

$$\langle\bar{n}_+\,|\,\overline{(n-q)}_-\rangle = J_q\left(\frac{\omega_1}{\omega}\right) \tag{III.9}$$

(J_q is the Bessel function of order q.)

By using this identity it is possible to compute exactly the intensity of the resonances as a function of ω_1/ω. Some experimental verifications of these calculations have been shown in Figure 11.

(5) *Modification of an atomic Landé g-factor by the coupling with a RF field* [19]. We return to the situation where $\vec{H}_1$ and $\vec{H}_0$ are perpendicular and we now focus our attention on the limit $H_1 \gg H_0$.

We want to study the energy diagram of $\mathscr{H}$ (Fig. 14) around the zero magnetic field value and determine the slopes of the crossing energy levels as a function of n (or ω_1).

We now consider $\omega_0 J_z$ as a small perturbation of the main hamiltonian

$$\mathscr{H}_0 = \omega a^\dagger a + \lambda J_x(a + a^\dagger).$$

Using the results of § III.B.4, the eigenstates and eigenvalues of $\mathscr{H}_0$ can be easily determined.

One finds

$$\mathscr{H}_0\,|\varepsilon\rangle_x|\,\bar{n}_\varepsilon\rangle = \left(n\omega - \frac{\lambda^2}{4\omega}\right)|\varepsilon\rangle_x|\bar{n}_\varepsilon\rangle$$

where $|\varepsilon\rangle_x$ are the eigenstates of J_x and $|\bar{n}_\varepsilon\rangle$ is given by (III.7). For each value of n, the states $|+\rangle_x\,|\bar{n}_+\rangle$ and $|-\rangle_x\,|\bar{n}_-\rangle$ have the same energy. We have a two-fold degeneracy, which is removed by the perturbation $\omega_0 J_z$. The new

energies are the eigenvalues of the matrix $\omega_0 J_z$ in this 2×2 multiplicity

$$\begin{pmatrix} 0 & \frac{\omega_0}{2} \langle \bar{n}_- \mid \bar{n}_+ \rangle \\ \frac{\omega_0}{2} \langle \bar{n}_+ \mid \bar{n}_- \rangle & 0 \end{pmatrix}$$

Using the relation (III.9), we easily find the two eigenvalues

$$\pm \frac{\omega_0}{2} J_0 \left(\frac{\omega_1}{\omega} \right).$$

The slopes of the two energy levels (i.e. the g-factor of the "dressed" atom) are thus different from those of the free atom by the factor $J_0(\omega_1/\omega)$:

$$\frac{g_{(\text{atom + RF field})}}{g_{(\text{free atom})}} = \frac{g_t}{g_0} = J_0 \left(\frac{\omega_1}{\omega} \right)$$

We have observed this effect experimentally. As we make a zero field crossing experiment (Hanle effect), we get a resonance whose width is inversely proportional to the slope of the crossing levels. As we increase H_1 (measured by the parameter V_1), the width of the resonance curve increases, becomes infinite for a certain value of H_1, decreases again and so on (Fig.15).

By measuring the variation of the inverse of the resonance width as a function of ω_1/ω, one can obtain g_t/g_0 as a function of the same variable.

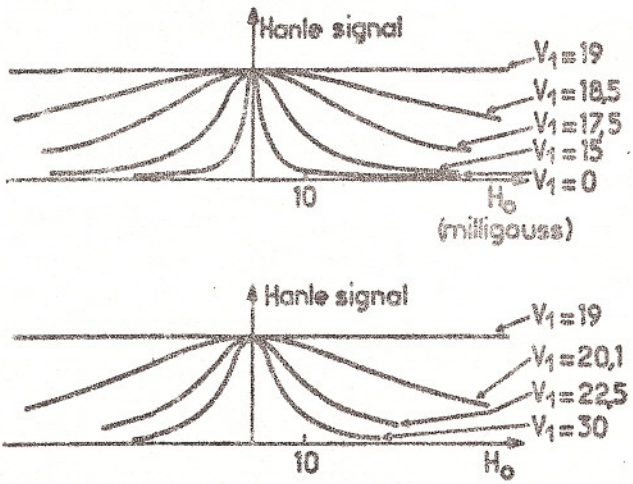


Fig. 15

The results obtained are in good agreement with the theoretical prediction represented by the full curve of Figure 16.

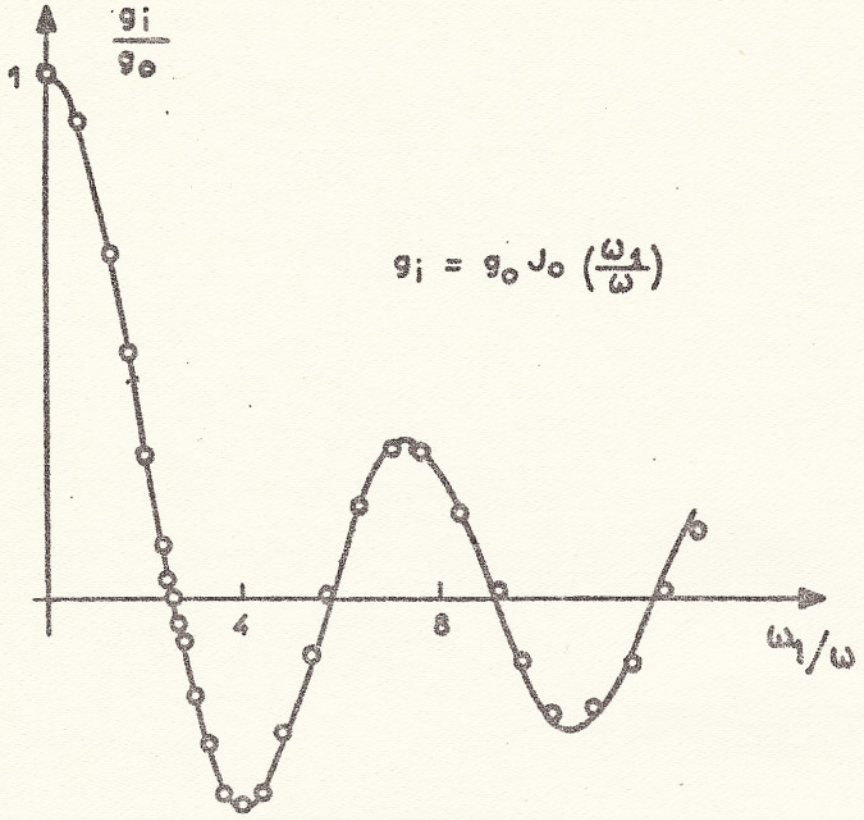


Fig. 16

Finally, all the experiments described in the chapter III have been explained within the same theoretical framework. Considering the atom dressed by the RF photons as a whole quantum system (S) has been very useful.

Conclusion

We will conclude with the following remark.

Several other courses of this session have been devoted to the study of the Lamb-shift and of $g - 2$. These two basic effects of Q.E.D. may be visualized as due to virtual emissions and reabsorptions of photons. We hope that we

have shown in this course that similar effects exist when the atom absorbs first and reemits impinging quanta: atomic levels can be shifted (light-shifts); atomic g-factors can be considerably modified.

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